Technology

Superconductors

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Fermi Surface Topology, Bilayer Splitting, and (π,0) dispersion kinks in Bi2212

A. D. Gromko[1],Y.-D. Chuang[1,2], A. V. Fedorov[1,2], J. D. Koralek[1], Y. Aiura[3], Y. Yamaguchi[3], K. Oka[3], Yoichi Ando[4], D. S. Dessau[1]

[1]Department of Physics, University of Colorado, Boulder, CO 80309-0390, USA
[2]Advanced Light Source, Lawrence Berkeley National Lab, Berkeley, CA 94720, USA
[3]National Institute of Advanced Industrial Science and Technology (AIST), AIST Tsukuba Central 2, 1-1-1 Umezono, Tsukuba, Ibaraki 305-8568, JAPAN
[4]Central Research Institute of Electric Power Industry (CRIEPI), 2-11-1 Iwato Kita, Komae, Tokyo 201-8511, JAPAN

INTRODUCTION

There have been many key developments in our understanding of the E vs. **k** band dispersion in $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) recently, both in the normal and superconducting states. Earlier work of ours [1] was the first to break tradition and indicate that the Fermi Surface (FS) of Bi2212 was not simply a single hole-like pocket centered around the (π,π) points of the Brillouin zone. In particular, we found that a more electron-like portion of the FS was observable at certain photon energies such as 33 eV, in contrast to the standard hole-like FS, which is observed at photon energies near 20 eV. Papers [2] and [3] dealt with this issue and principally contained data from the ALS. The concept of an electron-like FS portion in Bi2212 was supported by additional measurements made by the Stanford group [4]. This Fermi surface topology issue has evolved into the finding of bilayer split bands in the Bi2212 family [6,7]. This discovery explains the observance of two different FS topologies, and has allowed us to unearth new self-energy effects in the E vs. **k** band dispersion near the critical (π,0) point of the Brillouin zone [8,9].

EXPERIMENT

Experiments were carried out at beamline 10.0.1.1 of the Advanced Light Source, and at beamline 5-4 of the Stanford Synchrotron Radiation Laboratory using Scienta SES 200 electron spectrometers. Experiments were conducted at photon energies of 20, 22, 33, and 47eV. The measurements requiring the highest resolution were carried out with a combined experimental energy resolution of 12meV, and a momentum resolution better than $0.01\pi/a$ (where a is the CuO_2 plane lattice constant) along the entrance slit to the spectrometer. Experiments were carried out with the in-plane component of the photon polarization along the (0,0)-(π,0) direction.

RESULTS AND DISCUSSION

The bilayer splitting effect in Bi2212 is expected to occur due to the intracell c-axis coupling between the two CuO_2 layers per unit cell. However, this had not been previously observed, with most experiments indicating that the coupling was zero [5], a possibility consistent with exotic theories of superconductivity such as those favoring the low-dimensional state necessary for spin-charge separation. Simultaneous to the Stanford group's report [6], we made the first measurements of bilayer splitting in a high temperature superconductor, using overdoped Bi2212 samples [7]. We found the splitting to have a maximum value of approximately 100 meV, to be maximal at the (π,0) point of the Brillouin zone, and to be zero

along the (π,π) nodal direction. The large value of this splitting relative to other parameters such as the value of the superconducting gap, pseudogap, and some of the magnetic energy scales means that this intracell coupling is strong and should be included in any proper description of the electronic structure. This also should be necessary information for understanding why the T_c of the cuprates depends so strongly upon the number of layers per unit cell. Data showing this splitting is contained in figure 1.

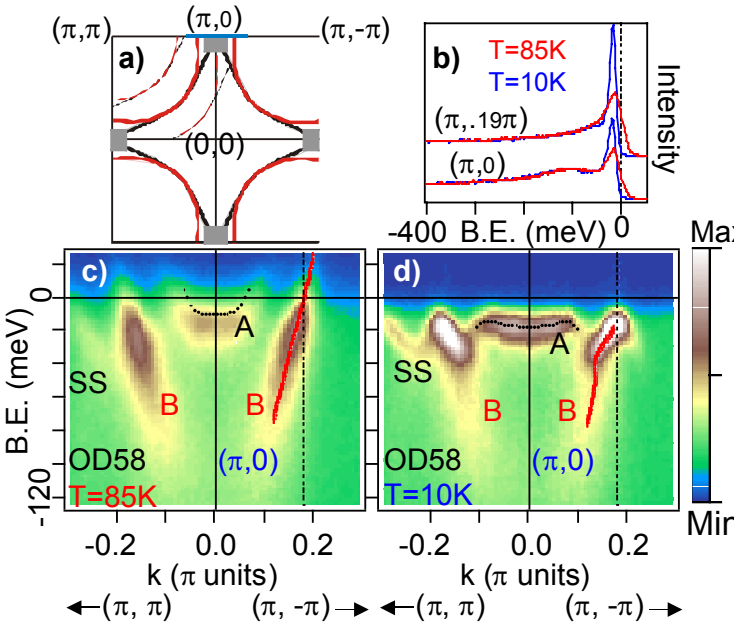

Figure 1. ARPES data in the normal (c) and superconducting state (d) of a T_c=58K overdoped Bi2212 sample, from ref [9]. A and B indicate the antibonding and bonding bands, respectively. The k-space location is along the blue line of panel (a). Panel (b) shows EDCs at two k-space locations.

A later paper of ours made the first extension of the bilayer splitting measurements to optimal and underdoped samples [8]. In these samples, the intrinsic peak broadening is greatly increased, and it is more difficult to separate the contributions of the separate bilayer split bands. By choosing optimal photon energy and polarization conditions we were able to selectively enhance one of the bilayer split bands relative to the other and make an accurate deconvolution. While the decreased c-axis conductivity in underdoped samples led us to expect that the bilayer splitting would be reduced in these samples as well, we found the surprising result that the bilayer splitting energetics are essentially identical for all doping levels [8]. This implies that other effects such as the scattering rate or the opening of the pseudogap must be more relevant to the change in the c-axis conductivity.

The ability to accurately deconvolve the bilayer splitting opened up many more opportunities for research in the critical (π,0) region of the Brillouin zone, where the bilayer splitting is the largest, as well as where the superconducting gap, pseudogap, van-Hove singularity, etc. are all maximal. Earlier measurements had been unknowingly looking at a superposition of the two bands in this region, with misinterpretations arising because of this. For example, there was the very famous peak-dip-hump lineshape at (π,0) observed by many groups in the superconducting state of Bi2212, but not in the normal state. This had been discussed as various types of self-energy effects, including $\alpha^2F(\omega)$ oscillations, coupling to magnetic modes, shake-off effects, etc. Our new data indicates that it is simply an effect of the bilayer splitting [9]. It is present in both the normal and superconducting states (figure 1b), but it was only previously visible in the superconducting state because the broad features present in the normal state pushed the signal below the background.

Even more importantly, the ability to accurately deconvolve the bilayer split bands has allowed us to make the first measurements of a dispersion "kink" near the (π,0) portion of the

Brillouin zone (figure 1d). This result follows up on the pioneering kink studies of cuprates done by the Stanford [10] and other [11,12] groups, except that these previous results were predominantly limited to the nodal region, where the pairing correlations are weakest. We have measured the temperature and momentum dependence of the new $(\pi,0)$ kink on over and optimally doped samples. We find that the kink strength (but not its energy scale) is a strong function of these parameters. In particular, the kink appears just below T_c, existing only in the superconducting state, while the nodal kink is present both above and below T_c. Also, the kink is localized in a small k-space region near $(\pi,0)$, with a momentum dependence that closely matches that of the famous "41 meV" magnetic resonance mode observed in inelastic neutron scattering measurements [13]. We argue that these factors point to a likely connection between the $(\pi,0)$ kink and the magnetic resonance mode, although more work needs to be done to understand the energy scales. If this picture holds together, the kink should be due to electronic coupling to the magnetic resonance and there will be a strong possibility that the pairing of electrons is mediated by this magnetic mode. This should be a very active and important area for future study.

ACKNOWLEDGEMENTS

We acknowledge sample preparation help from M. Varney, beamline support from X.J. Zhou, P. Bogdanov, Z. Hussain and D.H. Liu, and helpful discussions with G. Aeppli, A. Chubukov, C. Kendziora, A. Millis, P. Lee, D. Pines, D. Scalapino, J. Schmallian, Z-X. Shen, and S.C. Zhang. We gratefully acknowledge the help of R. Goldfarb at NIST for the use of the SQUID magnetometer.

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This work was supported by the NSF Career-DMR-9985492 and the DOE DE-FG03-00ER45809. ALS and SSRL are operated by the DOE, Office of Basic Energy Sciences.

Principal investigator: Dan Dessau, University of Colorado, Department of Physics, Boulder, CO 80309. Email: dessau@spot.colorado.edu. Telephone: 303-492-1607.

Infrared conductivity of photocarriers in organic molecular crystals

Chris Weber*[1,2], Ch. Kloc[3], Michael Martin[4], J.H. Schoen[3], B. Batlogg[3], Joe Orenstein[1,2]
[1] Department of Physics, University of California, Berkeley, CA 94720
[2] Material Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720
[3] Bell Laboratories, Lucent Technologies, Murray Hill, New Jersey 07974-0636
[4] Advanced Light Source Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720

This project, begun in March of 2001, seeks use infrared spectroscopy to probe the properties of photocarriers in organic molecular crystals. A group at Bell Laboratories has recently succeeded in creating FETs on ultrapure single crystals of pentacene, tetracene, and other "polyacenes" (rigid, rodlike chains of Benzene rings). Carriers injected into these transistors have exhibited metal-insulator transitions, superconductivity, lasing, and the quantum Hall effect (see, e.g., J. H. Schoen, Ch. Kloc, B. Batlogg, *Science* v. 288 p. 2338; *Science* v. 288 p. 656; *Nature* v. 406 p. 702). The goal of our experiment is to photoexcite carriers in these same materials, and to measure the infrared spectrum of these photocarriers.

The apparatus, at ALS beamline 1.4, includes an Argon-ion laser for photoexcitation of charge carriers, a Bruker 66v/S Fourrier-Transform Infrared (FTIR) spectrometer, and a variable-temperature cryostat. The laser allows photoexcitation at energies from 2.4 eV to 3.5 eV. At the UV frequencies, the quantum efficiency of photocarrier generation in pentacene is high, about 30%. The light is coupled to the sample (in the cryostat) by means of an optical fiber. The cryostat allows us to reach temperatures from 5 K to 300 K, and the FTIR spectrometer allows measurement of infrared transmission on the range of at least 100 cm^{-1} to 7800 cm^{-1}.

The basic result of a measurement is a transmission spectrum, $T(\omega)$, either with the sample illuminated with laser light, $T_{on}(\omega)$, or with the sample unilluminared, $T_{off}(\omega)$. From these spectra we determine difference spectra, $\Delta T / T = (T_{on}(\omega) - T_{off}(\omega)) / T_{off}(\omega)$. Our measurements at room temperature on crystals of Tetracene have shown good reproducibility, both in the shape and in the magnitude of the difference spectra. The measurement is sensitive to changes in the transmission of one part in 10^4 or better through most of the spectral range. We have resolved many clear and reproducible features that are an order of magnitude larger than the noise.

Although these results demonstrate that the apparatus is, indeed, able to make the desired measurements, they do not put us particularly close to our scientific goals. Most or all of the features we have seen thus far appear to be due to shifting or broadening of phonon absorptions due to the heating effect of the laser. This result at room temperature is no surprise, as the mobility of carriers in polyacene crystals increases as T^{-2} below room temperature, so that the conduction of the carriers should only become visible at low temperatures. More surprising was the result that tetracene crystals large enough to measure optically will invariably shatter upon cooling below about 180 K (the temperature seems to vary a bit). Pentacene crystals, on the other hand, do not shatter, but also do not seem to be available in sizes large enough for optical measurement. Other

polyacenes have much lower efficiencies of photocarrier generation (due to their having broader bandgaps), and so are not suited to this experiment.

We are now seeking thin-film samples of the polyacenes, as these should survive cooling and should have the large area desirable for infrared measurement. High-quality pentacene films have been shown to display many of the same electronic properties as their single-crystal counterparts (J. H. Schoen, Ch. Kloc, *Applied Physics Letters*, v. 79, p. 4043).

Funding sources: This work and the Advanced Light Source are supported by the Director, Office of Science, Office of Basic Energy Science, Material Science Division, of the United States Department of Energy under contract number DE-AC03-76SF00098 at Lawrence Berkeley National Laboratory.

* To whom correspondence should be addressed:
cpweber@lbl.gov
(510) 486-5879

Microstructure and pinning properties of hexagonal-disc shaped single crystalline MgB_2

C. U. Jung[1], J. Y. Kim[1], P. Chowdhury[1], Kijoon H. P. Kim[1], Sung-Ik Lee[1], N. Tamura[2], W. A. Caldwell[2], and J. R. Patel [2,3]

[1]National Creative Research Initiative Center for Superconductivity and Department of Physics, Pohang University of Science and Technology, Pohang 790-784, Republic of Korea

[2]Lawrence Berkeley National Laboratory, Advanced Light Source, 1 Cyclotron Road, MS-2-400 Berkeley, CA 94720, USA

[3] SSRL/SLAC, Stanford University, CA 94309, USA

INTRODUCTION

The existence of impurities and structural imperfections on a microscopic scale can result in diverse transport and pinning properties in MgB_2, which was even observed for single crystalline samples made by several groups[1-3]. Here, we report the X-ray micro-diffraction measurements for, MgB_2 single crystals with hexagonal-disc shapes and shiny surfaces. The diagonal length and the thickness for the largest crystal was about 100 μm and 10 μm, respectively. The crystallinity was thoroughly identified by using the Laue pattern in the X-ray micro-diffraction measurement. Both the edge and the c-axis of the hexagonally shape disc were found to match the crystal symmetry.

EXPERIMENTAL

Two different procedures were used to grow the single crystals, and in both cases, excess Mg was critical for the growth of single crystals. The first involved a two-step method in which already synthesized pieces of MgB_2 bulk[4] were used as a seed material. They were heat treated in a Mg flux inside a Nb tube, which was sealed in an inert gas atmosphere. Then, the Nb tube was put inside a quartz tube, which was sealed in vacuum. The quartz tube was heated for one hour at 1050 °C, cooled very slowly to 700 °C for five to fifteen days, and then quenched to room temperature. The crystal images were observed using a polarizing optical microscope and a field-emission scanning electron microscope (SEM). We successfully separated single-crystalline MgB_2 from the Mg flux by using a thermo-mechanical spinning method.

For the X-ray micro-diffraction measurements, several crystals were fixed at the center of Cu crosshairs on the substrate, as shown in Fig. 1. The instrument used at the Advanced Light Source (ALS) for X-ray micro-diffraction is capable of producing a submicron-size X-ray microbeam and with submicron spatial resolution can probe the local texture in a single crystal[5]. The sample was positioned using the Cu fluorescence signal detected from the Cu crosshairs on the Si substrate by using a high-purity Ge ORTEC solid-state detector connected to a multichannel analyzer. The crystal orientation with respect to the substrate can be determined with an accuracy of 0.01 degree.

RESULTS

The crystal structure was identified by using white beam X-ray micro-diffraction measurements. After positioning these single crystals, a 100 μm × 100 μm region between the Cu crosshairs was scanned with a step size of 2 μm. At each step, the Laue pattern (together with the Cu *K*

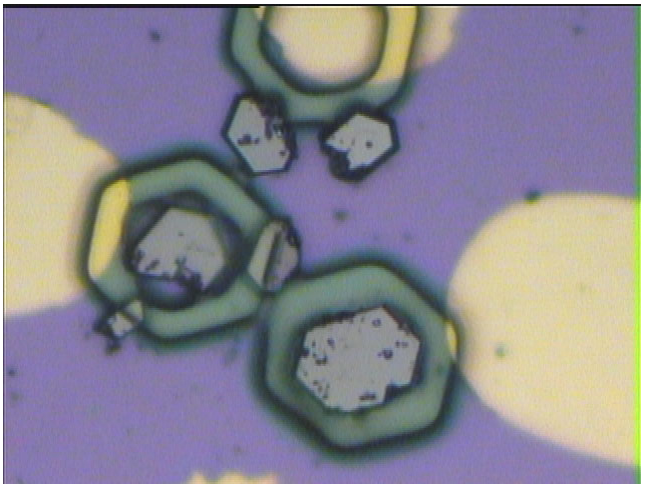

Figure 1. Polarizing optical microscope images of MgB_2 single crystals. An epoxy was used to fix six single crystals at the center of 100 μm-wide Cu crosshairs.

fluorescence signal) was collected with a BRUKER 6000 CCD camera which has an active area of 9 × 9 cm and was placed about 4 cm above the sample. [2500 images, 1024 pixel ×1024 pixel mode] The exposure time at each step, was 1 second. An example of a Laue pattern obtained from a MgB_2 single crystal is shown in Fig. 2(a). The Laue patterns are consistent with a hexagonal MgB_2 structure (a =0.3086 nm, c = 0.3524 nm, Space Group number =191, Ref. 6). The (0005) reflection in the center of the pattern in Fig. 2(a) corresponds to the direction of the normal to the crystal surface. This confirms that the surface plane normal is along the c-axis. Moreover, the hexagonal edges of the crystals were found to match the <1,0,-1,0> directions within a fraction of a degree resolution. Thus, the shapes of the crystals in the microscope image followed the MgB_2 crystal symmetry, which will be quite useful for any research of the direction dependencies of the physical properties in MgB_2.

Indexing the Laue patterns in Fig. 2(a) allowed us to calculate the complete orientation matrix of the X-ray illuminated volume. A finer step size of 1 μm was used for the white-beam scan. The orientation variations inside the single crystal shown in the right bottom corner of Fig. 1 are shown in Figs. 2(b) and 2(c). Figure 2(b) is the out-of-plane orientation variation calculated as the angle between the c-axis and the normal to the surface of the silicon substrate. The out-of-plane variation was about 0.2 degrees between the light orange and the red regions. The out-of-plane orientation shows a variation of about 0.2 degrees between the bottom and the top parts, indicating a slight bending of the crystal (which might be due to the photoresist used as an epoxy). Figure 2(c) shows the in-plane orientation variation calculated as the angle between the measured a-axis (or b-axis) and a reference directions. The variation was about 0.4 degrees between the light blue-green and the green regions. The in-plane orientation also showed some inhomogeneities of up to about 0.2 degrees.

These results demonstrate that the orientation of crystal axis of our hexagonal-disc-shaped single crystals was perfect, within 0.2 degrees[7]. A recent study showed that (0001) twist grain-boundaries, formed by rotations along the c-axis (typically by about 4 degrees), were the major grain boundaries in polycrystalline MgB_2 [8].

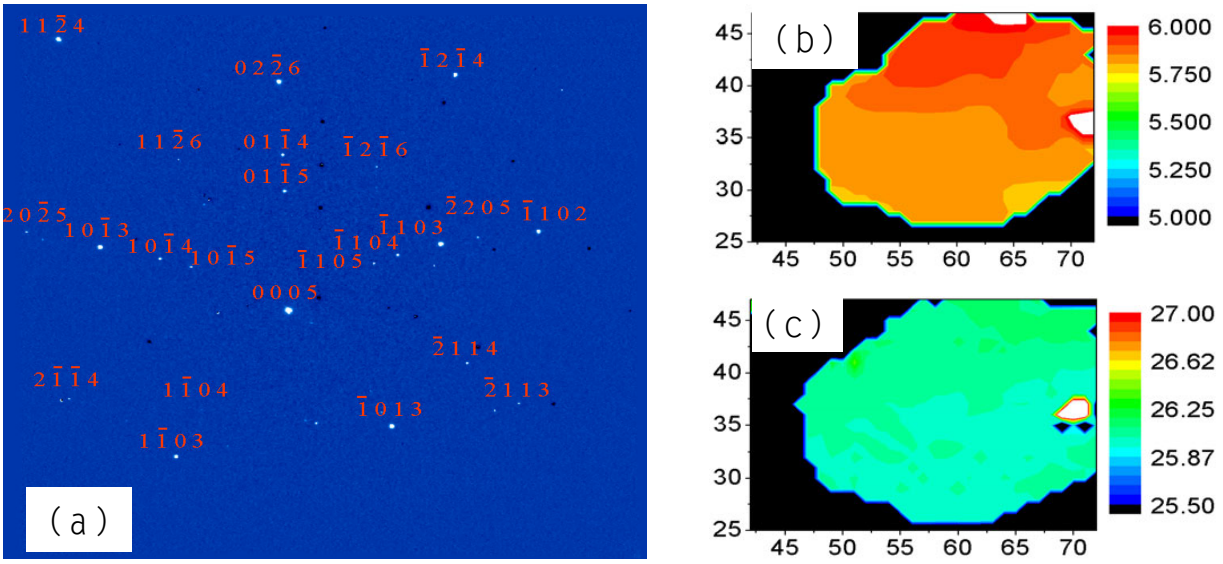

Figure 2. (a) A representative image of an indexed Laue pattern from X-ray micro-diffraction. (b) and (c) are, respectively, for the out-of-plane and the in-plane orientations inside a single crystal.

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The work done at the Advanced Light Source is supported by the Director, Office of Science, Office of Basic Energy Sciences, Materials Sciences Division, of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098 at Lawrence Berkeley National Laboratory. Part of work done at PSC is supported by the Ministry of Science and Technology of Korea through the Creative Research Initiative Program.

Principal investigator: Nobumichi Tamura, Advanced Light Source, Ernest Orlando Lawrence Berkeley National Laboratory. Email: ntamura@lbl.gov. Telephone: 510-486-6189.